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Methodology for Determining Radionuclide Concentration in Groundwater in the Vicinity of Accelerator and Beamline Enclosures

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July 2002

Abstract

A method for estimating concentrations of radionuclides in groundwater due to operation of an accelerator facility, taking into account the velocity of the groundwater in the vicinity of the activation region is described. This methodology is an extension of the Fermilab Concentration model, motivated by the design of the NuMI Facility which lies predominantly in the aquifer of the Silurian Bedrock. This methodology has been reviewed and accepted by the Fermilab ES&H Section for application to the design, construction and operation of the NuMI facility.

Introduction

Operation of a particle accelerator and its associated external beamlines has the potential to activate soil, rock and water in the vicinity of the beam enclosures. At the Fermilab accelerator complex potential groundwater irradiation is calculated using a model which takes into account the source of the irradiation, the accelerator or beamline geometry and the surrounding geological environment. In the past, two different models have been used, namely, the Single Resident Well Model and the Concentration Model (Ma93),(C099-1).

In 1997 design work began on the NuMI Project which involves construction of a kilometer long external beamline which is at an angle of 54 milliradians with respect to the plane of the accelerator tunnel (TDR99). The NuMI Beam uses 120 GeV protons extracted from the Fermilab Main Injector to produce a secondary hadron beam and a tertiary neutrino beam. Proton intensities up to 4×10^{13} per cycle (~2 sec) are required to create a sufficient neutrino flux to carry out the physics program and the unprecedented intensity added many new challenges into the facility design.

The pitch to the beamline causes it to traverse several different geological media, including the Silurian dolomite which is classified as a

Class 1 groundwater aquifer[1]. Because the earlier models used by the Laboratory did not address this beamline geometry or geological region, a new methodology had to be developed for the NuMI Project. The purpose of this Technical Memo is to describe this methodology. Application of the

[1] A Class ! groundwater aquifer is defined in Illinois Administrative Code Environmental Regulations, Title 35 as ....

methodology and the parameters which have been used in the calculations

Specific calculations require modeling of the groundwater transport through the various geological media surrounding the facility enclosures, the primary beamline transport and the production and transport of the secondary beam, and finally, radionuclide production and decay. In the following sections the motivation for, and the general principles applying to each of these topics is discussed.

Hydrogeology at the Fermilab Site

As mentioned above, prior to the NuMI Project the only site geology considered for construction projects was the Glacial Till which extends to a depth of approximately 60 feet. The till is predominantly over-consolidated clay, sand/silt lenses and organics. Though the soil in this region can be quite wet, even saturated, the water is not considered to be a groundwater resource because it moves very slowly (cm's per year). Calculations of potential irradiation make a reasonable assumption that activation can build up to saturation levels. The slow flow rate then allows for the decay in transit of radionuclides as the water moves toward the underlying aquifer. Enclosures in this region are generally constructed with a lining to prevent inward seepage of water.

Construction deeper than the Glacial Till involves two regions. The first is a transitional interface region of approximately ten feet in depth. The glacial till/ rock interface is an extremely variable region. The lowest most subglacial unit, the Batestown, is highly variable (coarse to fine-grained) in its composition or may be nonexistent. There is always a chance of outwash deposits (very coarse) at the interface of the glacial deposits and bedrock, and the nature of the upper dolomite can be from hard and competent to highly weathered. The water flow in the soil is extremely slow and the water flow in the dolomitic rock is several orders of magnitude greater. In general the aquifer begins about 10 to 15 feet above the interface region, depending on the occurrence of the Batestown and outwash deposits.

Beneath the interface region is the Silurian Dolomite which extends to a depth of about three hundred feet. Since groundwater classifications are based on the flow rates of the water in the region, the aquifer of concern to Fermilab construction projects begins in the interface region. No longer can credit be taken for "decay in transit" since the enclosure is located in the aquifer.

Groundwater calculations for beamlines traversing these two regions must take into account whether or not the enclosure is lined or unlined, respectively preventing or allowing the groundwater flow into the enclosure.

For lined tunnels one can conservatively assume that the water flows in the vicinity of the enclosure follows the regional formational dip to the southeast or toward an induced flow location like Well 1. For an unlined enclosure, if the water flows into it rapidly and is pumped out to surface waters, activity will not build up in the aquifer.

Average inflow velocities can be calculated, based on estimated inflow rates, tunnel geometry and grouting criteria. As will be seen in the methodology presented below, these velocities play a major role in estimating the resulting activation levels in the water flowing into an unlined tunnel. They can also be used to estimate the activation in water flowing out of the region around a lined tunnel.

Based on the recent tunneling experience in the Chicago area, groundwater inflow into rock tunnels is controlled by the secondary permeability of the rock mass, i.e. the near vertical joints and to a lesser extent the bedding planes. The magnitude of the steady state groundwater inflow in TARP[2] rock excavations averages around 0.022 to 0.04 gpm per lineal foot of tunnel. The methodology for calculating expected flow velocities based on local measured rock parameters is described in detail later.

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[2] TARP - define

Modeling of the Beamline

The primary beam transport and the production of the secondary beam is done using the MARS simulation program (Mo98). The user input to the program includes the parameters of the proton beam, namely energy, spot size and divergence, the layout of the beamline elements and their specific geometries, and the geometry and composition of shielding and enclosures surrounding the beam elements. The user can also set various thresholds which determine the level to which particles are tracked, depending on the specific calculation which is desired. Output from the MARS simulation includes particle fluxes, star densities[3], energy deposition and residual dose rates. For the purposes of groundwater calculations only the star density produced in the soil or rock surrounding the beamline enclosures is relevant. It has been shown that star production falls exponentially away from the enclosure walls allowing calculations to limit the volume of the soil or rock which must be modeled. The methodology presented in this memo uses a "99% volume" (to be defined in detail later), which typically extends about two meters into the soil or rock surrounding the beam enclosure. Star density produced by a simulation of the beamline operation can be used to predict a radionuclide concentration.

Radionuclide Production and Regulatory Requirements

The production of radionuclides in soil, rock or water surrounding an operating accelerator or beamline is dependent on the beam parameters such as energy and particle type and on the chemical composition of the soil or rock. Many years of studies have shown that there are only two radionuclides of concern produced in the vicinity of the Fermilab accelerator complex, namely ^{3}H and ^{22}Na (Bo72).

The standards for groundwater (DOE Order 5400.5 and 40 CFR 141) are designed to limit doses to the public to 4 mrem per year. The current regulatory limits for both groundwater and surface water discharge for ^{3}H and ^{22}Na are summarized in **Table 1**. The sum of the fractions of radionuclide contamination (relative to the regulatory limits) must be less than one for all radionuclides;

$$\sum_i \frac{C_i}{C_{reg\ i}} \leq 1,$$

where the sum is over radionuclides, i, C_i is the concentration of radionuclide i in the water and $C_{reg\ i}$ is the regulatory limit concentration.

[3] A star is defined as a nuclear interaction above a threhold of 50 MeV. Star density is given as stars per cm3-proton.

Fermilab ES&H policy requires that a facility design must demonstrate that beamline operation will *not* result in activation levels above the regulatory limits, including all uncertainties in the methodology and input parameters. Verification that such limits are not violated is accomplished during the facility operation through the Lab-wide monitoring program (FESHM).

Methodology

The concentration of a radionuclide, *i*, in groundwater in the vicinity of a beamline enclosure is given by the following equation (Cos99-2):

$$C_i\left(\frac{pCi}{ml}\right) = R_{till}\frac{N_p S_{avg} F_i}{0.037}\left[\frac{\lambda_i}{\left(\lambda_i + \frac{v_i}{r}\right)}(1 - e^{-\left(\lambda_i + \frac{v_i}{r}\right)t_{ir}})\right] \tag{1}$$

where

$$F_i\left(\frac{atoms}{star}\right) = \frac{K_i L_i}{n}\,. \tag{2}$$

The factor (1/0.037) is the conversion factor used to convert disintegrations per second (dps) to picoCuries, and the term in brackets is the buildup/removal factor, accounting for radionuclide buildup, decay and removal due to water flow. The factors in the equation are defined and then discussed in more detail below. The first factor is simply a given constant. The next three are determined by operating scenarios and the model of the facility using the MARS simulation program. The values used for these numbers depend somewhat on the hydrogeologic conditions. The remainder depend completely on the hydrogeology in which the facility is located. It is important to note that for extended facilities such as the NuMI enclosures and tunnels, these parameters may vary along the facility and thus concentrations must be calculated in a region dependent manner.

λ_i is the inverse mean lifetime of radionuclide i, measured in units consistent with those of time t_{ir}.

N_p is the number of incident protons per second at the source (protons/sec).

S_{avg} is the average star density per incident proton in a region of unprotected rock/soil close to the source of production (stars/cm^3/proton).

r is the radial distance from the tunnel wall that defines the volume over which the star density is averaged to determine S_{avg}.

$K_i L_i$ is the atoms per star for isotope i that is in the water (atoms/star).

n is the porosity of the rock formation; that is the ratio of the volume of void in the rock/soil (generally filled with water) to the volume of rock (unitless).

t_{ir} is the irradiation time (residency time in activation volume for flowing water).

v_i is the flow velocity for radionuclide *i* in water, which is directly dependent on the velocity of the groundwater, *v*, in the region of interest

R_{till} takes into account credit for decay and dispersion in transit to the acquifer. It is 1 for tunnels located in the aquifer and varies for glacial till.

The Inverse Mean Lifetime, λ_i

The mean life times for the two radionuclides of concern are 17.5 yr for ^3H and 3.7 yr for ^{22}Na.

The Number of Protons, Np

Typically the proton intensity is expressed in protons per second, taking into account the average number of protons extracted per cycle, as well as an overall efficiency factor for operation of the beam line. For static water, the value of N_p chosen should be representative of the average annual proton delivery. Given the nature of the Fermilab operations cycle, it is recommended that this average be taken over a three-year period. Assume an expected average “dc” rate given in protons per second. This corresponds to an expected number of protons/year.

For the accident cases, where one assumes only a few pulses, N_p should be the maximum protons/sec. Similarly, for cases where the residency time of the water in the activation region is less than or equal to 1 week, the maximum value of protons/sec should be used. For all other cases the average "dc" rate of protons per second should be used.

The Average Star Density, S_{avg}

In order to predict an activation concentration, one has to define a volume over which to sum activity. Figure 1 shows the drop off of star density radially in rock outside a thickly shielded beamline. The standard concentration model (Co99, Ma93) used the volume around the target or absorber that included 93% of the activation. The 93% of the activity corresponds to a volume where the star density has fallen to 1% of its maximum value and is called the “99%” volume. The average star density can be obtained from star density data, by going “out” from the maximum star density S_{max} to those radial and longitudinal values at which S has dropped to 1% of its maximum value.

For a uniform beam loss along a long tunnel, a cylindrical volume is reasonable to use. In general, as seen from Figure 1, two meters away from the tunnel wall, the star density has dropped to 0.1% of the maximum. One can also determine from this curve what radial distance would correspond to the 99% volume. For asymmetric geometries either in beam loss or tunnel geometry, one needs to check the star density fall off in different locations and determine the appropriate volume over which to sum. S_{max} is generally determined by creating 1cm radius radial bins in the region just outside the tunnel. For uniform loss, the beamline z dimension of these bins will be driven by the length of the loss, the tunnel geometry, or geological variations.

For non-uniform beam loss, as in an accident condition, the determination of S_{avg} is more difficult to determine. In flowing water regions, it is not very critical. It can be shown that the radionuclide concentration for water flowing at velocities encountered at the NuMI site is relatively independent of the star density volume, and thus it remains valid to use the 99% volume for the S_{avg} calculation. (For groundwater velocities significantly varying from these or for very non-standard geometries, one should check that this is true before assuming the 99% volume.) A plot of the sum of the relative concentration for the two radionuclides of concern, ^{3}H and ^{22}Na, versus activation volume is shown Figure 2 for several water velocities. The larger the volume one sums over to determine S_{avg}, the longer it takes the water in that volume to flow in to the tunnel. These two effects tend to cancel each other as the larger the volume one sums over, the smaller

S_{avg} becomes, and the longer the irradiation time becomes for the larger radius volume.

The Hydrogeology Factor, F_i

Since the writing of TM-2009 in the summer of 1997, Wehmann and Childress (WC99) have refined the values of the quantities that make up the factor F_i. For ^3H and ^{22}Na, this methodology uses the suggestion of Wehmann and Childress, who base their results on the measurements of Borak, et al (Bo72). Since the value of KL for tritium is based on actual measurements, it includes both the effects of production in the dolomite with subsequent entrapment in the water, as well as the direct production of tritium in the water in the rock itself. For ^{22}Na, direct production in the water is not possible.

Borak et al found that, at least for the case of tritium, "leachable" activity is associated with the amount of water in the soil or rock at the time of irradiation. Since the transport of radionuclides through the dolomite occurs under conditions in which the medium is saturated with water, it is the water in the voids that is available for "leaching". Porosity, n, is defined as the fraction (by volume) of the rock that is void of material.

From the report of Wehmann and Childress (WC99),

$$K_j = \left[\frac{\Sigma_i n_i \sigma_{ij}}{\Sigma_i n_i \sigma_{inel}}\right]\left[\frac{f_{new\_material}}{f_{Borak\_soil}}\right]$$

(3)

Where the first term in brackets is the average of Borak's measured cross sections for radionuclide *i* (tritium in the report) production in water in glacial till divided by the total inelastic cross section[4]. The second term is the percentage of water in the new material (rock or soil) divided by the average percentage of water in Borak's samples (0.132). Since

$$f = \frac{n}{\rho} \qquad \text{and} \qquad F = \frac{KL}{n},$$

then, in general (equation 3),

$$Fj = [\Sigma_i\ n_i\ \sigma ij\ /\Sigma_i\ n_i\ \sigma_{inel}][Lj/(f_{\ Borak\_soil})(\rho_{new\_material})]$$

$\rho_{new\_material}$ is the density of the new material (dolomite is 2.7 g/cm^3). K, the production probability, is based on production of activation by hadrons above 30 MeV, from the work of Borak, et al. On the other hand, the star density S in MARS is defined to be those interactions above a threshold of 47 MeV. MARS calculations gave a relationship between hadronic flux above 47 MeV

[4] WC99 also looked at the difference between the average cross sections for tritium production in dolomite versus glacial till and found them negligible.

and the hadronic flux above 30 MeV to be ~1.24. Therefore, for consistency with Borak, the value of F given above is multiplied by a factor of 1.24.

L is a "leaching" parameter that indicates the amount of radionuclide that is available to the water. Typically L is thought of as the amount of radionuclide that can be leached from the rock or soil into the water. For tritium in dolomite, L is assumed to be one, since all the tritium that is produced in the rock and water gets into the water. Thus, for example, F for tritium in dolomite is estimated as,

$$F_{^3H} = \frac{6.9 \times 10^{-4}}{1.1 \times 10^{-2}} \cdot \frac{1.24}{(0.132)(2.7)} = 0.22 \; atoms/star.$$

For ^{22}Na in dolomite, the values of L range from 1% to 2% (Cu00); and one uses an average value of 1.5%. Similarly, for ^{22}Na in dolomite, using Borak's average measured cross section[5],

$$F_{^{22}Na} = \frac{2.0 \times 10^{-4}}{1.1 \times 10^{-2}} \cdot \frac{(1.24)(0.015)}{(0.132)(2.7)} = 0.00095 \; atoms/star.$$

Equation (3) can be used in general for other materials of similar chemical composition.

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[5] Cu00 measured values of ^{22}Na production in water in NuMI dolomite (KL) of 0.0019 with 60% uncertainty. This agrees well with the value of 0.000143 extrapolated from Bo72 using WC99.

The Buildup/Removal Factor

The buildup/removal factor is a function of t_{ir}, v_i and r (see Equation 1). The distance, r, is the approximate distance one must go out from the tunnel wall radially to obtain an S_{avg} over the "99%" volume. A "generic" plot of star density versus radial distance from a tunnel center with thick shielding was shown in Figure 1.

The variable with the greatest effect on the buildup/removal factor is v_i, the velocity of radionuclide *i* as it travels within the water. ^{3}H flows at the same rate as the water, and the tritium produced in the rock, that is available to the water, is quickly picked up by the water. On the other hand, ^{22}Na is exchanged back and forth with the rock formation, thus retarding its flow rate. Based on Borak et.al, one can define:

$$v(^{22}Na) = \frac{v(water)}{1 + \frac{K_d \rho}{n}} = 0.21\, v(water)$$

Here K_d is the distribution coefficient for ^{22}Na that takes into account the ion exchange and thus the retarded flow rate. Borak measured K_d as 0.204 ml/g for Fermilab glacial till. K_d is determined by cation exchange and depends on the amount of clay in the material and the surface area available. We have not found any measurements for dolomite. The amount of clay in Fermilab dolomite is comparable to the amount of clay in Fermilab glacial till. The surface area available for exchange is less in dolomite than in glacial till, thus the glacial till value of Borak is conservatively used as an upper limit on K_d in dolomite. The average density and porosity for the NuMI dolomitic formations is 2.7 g/cm3 and 0.15, respectively. This gives a ^{22}Na velocity 0.21 times that of the water in the dolomite.

Irradiation time, t_{ir}

The irradiation time is a function of the residency time of the water in the activation region (99% volume for example). Thus,

$$t_{ir}\left({}^{3}H\right)=\frac{r}{v(water)};\quad t_{ir}\left({}^{22}Na\right)=\frac{r}{0.21\,v(water)}.$$

For regions where the velocity of the water is zero (glacial till), the irradiation time is based on the lifetime of the facility.

For ***v(water)*** one determines a weighted average velocity of the water flowing into the tunnel. This is a water volume weighted average of the water velocity through the matrix and the fractures. Two steps are needed to determine this weighted average velocity. First a fracture spacing is determined using the estimated inflow rates for each region and the inflow rate versus fracture spacing. Then the matrix and fracture flow velocities are weighted to determine the average velocity, using the fraction of inflow from the matrix versus fracture spacing data.

The weighted average velocity is calculated according to the following relationship:

$$v = \frac{1}{\frac{f_{matrix}}{v_{matrix}} + \frac{f_{fractures}}{v_{fractures}}}$$

where $f_{\text{matrix(fractures)}}$ is the fraction of water through the matrix (fractures) as det*v*rmined for each individual application and $\nu_{\text{matrix(fractures)}}$ is the velocity of the water through the matrix (fractures) in ft/year.

Decay in Transit, R_{till}

$\mathbf{R_{till}}$ is the variable that has been used to quantify the decay and dispersion in transit to the aquifer. It is highly recommended that one measure the conductivity in the region where activation is to occur and then use PATCH3 modeling software to estimate R_{till}. For the usual concentration model with R_{till}, the long half-life of tritium makes tritium the dominant concern

Determination of Uncertainty

Uncertainties must be reasonably estimated for all input parameters to this model and their effects on the final result shown as an error bar on that result. Table *2* summarizes the input parameters which require an uncertainty when calculating the result.

Summary

A methodology for estimating radionuclide concentrations in groundwater, has been presented. This methodology incorporates the velocity of groundwater flowing in the vicinity of the source of activation. By determining an upper limit to the concentration we have shown how a facility design can be compared to government groundwater standards. By incorporating water flow, measurably activated water reaching a well is a negligible concern for two reasons. Water that is flowing quickly, and thus will get to a well quickly, will flow quickly through the activation region and thus will not be activated significantly. Water which flows slowly will get more activated, but it will also flow slowly to a well and thus will decay, disperse and dilute greatly in transit. It should be noted that there is a medium range of flow velocity that is more of a concern, but still provides significant decay in transit to a well.

Table 1: Regulatory Limits for ^{3}H and ^{22}Na

Radionuclide	Groundwater Limit (pCi/ml)	Surface Water Limit (pCi/ml)
^{3}H	20	2000
^{22}Na	0.4	10

Table 2 Summary of uncertainties to consider in applying methodology

Uncertainty in	Can be estimated by :
N_p	NA; should design for reasonable intensity goal
S_{avg}	Statistical error in simulation jobs;should always be less than 10%
Hydrogeology factor Fi	Needs to be determined from the available measurements
Water velocity, vi	Needs to be determined from the available measurements

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